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THE ROUGH SEMICONDUCTOR ELECTRODE; A NEW PERCOLATION PROBLEM

By

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We show that current flow through a rough interface with an exhaustion layer of controllable width can be idealized into a novel percolation problem. For an infinitely rough interface between two perfect conductors and a layer of finite conductivity separating the two, there exists a thickness t^* such that the normalized resistance $R = 0$ for $t < t^*$, $R\alpha(t - t^*)^s$ for $t \to t^*$ and $R\alpha t$ for $t >> t^*$, where s is a universal constant that will vary only with the topology of the interface. We formulate the scaling relations for a real interface and for the practical solution where the separating layer is an exhaustion

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Abstract

We show that current flow through a rough interface with an exhaustion layer of controllable width can be idealized into a novel percolation problem. For an infinitely rough interface between two perfect conductors and a layer of finite conductivity separating the two, there exists a thickness t^* such that the normalized resistance $R = 0$ for $t < t^*$, $R \alpha (t - t^*)^s$ for $t \to t^*$ and $R \alpha t$ for $t \gg t^*$, where s is a unversal constant that will vary only wih the topology of the interface. We formulate the scaling relations for a real interface and for the practical situation where the separating layer is an exhaustion layer of a semiconductor and where t can be controlled by an applied voltage.

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We have recently reported evidence for three-dimensional percolation through the interface of a polycrystalline electrode (CdSe) with an electrolyte ($S^{=}$/S/NaOH in H_2O).[1] Scaling behavior was observed in the frequency dependence of both the differential resistance and the differential capacitance of the interface. The exponents were, within experimental error, those expected in the vicinity of a three-dimensional percolation transition. The range of frequencies for which the scaling behavior occurred increased towards lower frequencies as the d.c. voltage across the electrode approached what appeared to be a percolation threshold. The d.c. current-voltage characteristics did not, however, show evidence of a sharp percolation transition.

In our previous report, we attributed this behavior to roughness of the semiconductor-electrolyte interface. In particular, the interface of the electrodes we studied had a cauliflower-like structure interpenetrated by the electrolyte.[2] In such a structure, all current flowing from the electrolyte into the bulk of the semiconductor electrode must pass through an exhaustion layer at the interface. Once through the exhaustion layer, it passes through the low resistance semiconducting material within the cauliflower structure and ultimately into the bulk of the semiconducting electrode. However, as the d.c. voltage moves away from the flatband potential, the exhaustion layer grows further into the interior of the cauliflower structure. The low-resistance paths are successively pinched off at constrictions until something like a percolation threshold is reached. Subsequent to that the current flows through the interface primarily in the regions around the base of the stems. The effective area through which current flows from the electrolyte into the semiconductor thus becomes of order of the geometric area of the interface instead of the actual area as was the case before pinch-off started. The resistance thus increases substantially.[3]

These phenomena are significant beyond the AC characteristics of liquid-junction solar cells. We expect such behavior to be most prominent in electrochemical storage systems in which the formation of the interface exhaustion layer is controlled by the state of charge of the storage electrode and by the potential drop across the interface. In general, all porous or rough semiconductor electrodes should exhibit similar behavior.[4]

In the present paper, we formalize these notions, which were largely implicit in our previous paper. We show that current flow through a rough interface with an exhaustion layer of controllable width can be idealized into a novel percolation problem. The deviations from the ideal case can then be embraced by a set of scaling relations.

The idealized percolation problem is defined as follows. Consider two perfect conductors, A and B, separated by a rough interface S with random aspects to its geometry. Introduce cartesian coordinates x, y, z. At the system boundary $z = Z/2$, space is occupied solely by A, and at the boundary $z = -Z/2$ solely by B. We suppose that the interface is, on average, planar. That is, if we define the interface by specifying the random function $z_i(x,y)$, then the ensemble average of $z_i(x,y)$ is zero. The average position of the interface coincides with the x-y plane. We now suppose that z_i is a bounded function of x and y. That is, the plane $z = z_{max}$ bounds the interface on the A side, and $z = z_{min}$ bounds it on the B side, $L = z_{max}-z_{min}$, cf Fig. 1. We choose as the simplest measure of the roughness of the interface its specific area $\mathcal{S}$, that is, the ratio of the actual area F of the interface to the cross section of the system perpendicular to the z-axis, the geometric area G.

$$\mathcal{S} = F/G \tag{1}$$

We now discuss explicitly the resistance of the system. This is introduced by supposing that a layer of material C of resistivity ρ and thickness t is allowed to grow out from the interface within the space originally occupied by the perfect conductor B, as shown in Fig. 1. The product $\mathcal{R}$ of the total resistance R and the geometric area G of the interface is linear in t for sufficiently small t,

$$\mathcal{R} = RG = \rho t/S. \tag{2}$$

As t increases, some of the zero resistance channels from portions of the interface to the bulk semiconductor are interrupted by regions of finite resistivity ρ so that the effective interface area decreases, increasing $\mathcal{R}$. This process increases with increasing t until ultimately all such paths are closed off, and R becomes asymptotically proportional again to t and inversely proportional to G

$$R = \rho t/G, \ \mathcal{R} = \rho t. \tag{3}$$

This dependence of $\mathcal{R}$ on t is sketched in Fig. 2.

Now suppose that the interface becomes infinitely rough, which can occur in two ways. First, both L, the thickness of the interface, and Z, the extent of the system along the z-axis, can diverge. However, for the system to retain its significance as an idealization of a rough electrode in an electrolyte, we require that

$$\lim_{\substack{Z\to\infty \\ L\to\infty}} L/Z = 0 \tag{4}$$

Second, the distance scale λ on which $z_i(x,y)$ fluctuates can go to zero while L remains finite so that $L/\lambda \to \infty$. In the latter case, there would be no space to replace a layer of B of finite thickness at S by C, and we could not construct in this way an idealization of a rough semiconductor electrode. Accordingly, we choose the first case in which S goes to ∞ while λ remains finite, so that $L/\lambda \to \infty$ only as $L \to \infty$.

Consider now the behavior of R with t in the limit that $S \to \infty$ as above. R and $\partial R/\partial t$ are zero at t = o. For there to be a true percolation threshold at some t > o, however, R must remain zero for all t, $o \leq t \leq t^*$. The process of erosion of B at and inside S and its replacement by C defines a mapping of S onto the interface I between C and B. Any point P_S on S will be mapped thereby into a point P_I on I. If the measure of points P_S for which there can be drawn a continuous path through B from the corresponding P_I on I to points $z < z_{min}$ is infinite, R will remain zero. Such a path cannot be drawn if P_I is on a disjoint piece of I. As long as t is sufficiently less than λ, the probability that P_I is on a disjoint piece will be less than unity. As t increases to t*, however, that probability becomes unity. For larger t, the effective area of the electrode becomes of order G and R becomes finite. To summarize,

$$R = o, \quad o < t < t^*$$
$$R = \rho f(t) > o, \quad t^* < t, \tag{5}$$

as shown in Fig. 2.

Asymptotically, we expect[5]

$$f(t) \sim K(t-t^*)^s \qquad t \to (t^*)^+$$
$$\sim t \qquad \frac{t}{L} \to \infty, \frac{t}{Z} \to 0 \tag{6}$$

For a suitably multiply-connected interface, the exponent s has the universal value expected in a three-dimensional percolation processs, 1.6, and K is a universal constant.[6,7] If the interface is less complex, the value of s may differ. In the limiting case of a simply connected surface, s may go to zero and there may be a step in f at t*.

The real interface differs from the above idealization in several ways. First, the electrolyte A has finite resistance ρ_A, the bulk semiconductor has finite resistance ρ_B, and the specific area S is also finite. Second, the exhaustion layer C at the interface is not a simple ohmic material of resistivity ρ but is highly nonohmic. We deal now with the first set of differences.

We allow S to become finite, keeping $\rho_\alpha = o$, α = A or B. R is then given by

$$R = \rho g(t, t/S)$$

for all t. The function g has the limiting behaviors

$$\begin{aligned} &g(t,o) = f(t) \\ &g(t,\tfrac{t}{S}) \to \tfrac{t}{S},\ t/\lambda \to o \\ &g(t^*,\tfrac{t^*}{S}) = \beta S^{-x},\ S \to \infty, \end{aligned} \tag{7}$$

where β is a nonuniversal constant and x is a universal exponent. We next allow ρ_α to become finite, α = A or B. We must now define R as

$$\mathcal{R} \equiv \mathcal{R}_{\text{Total}} - \frac{1}{2} \quad \rho_\alpha Z, \tag{8}$$

where $\mathcal{R}_{\text{Total}}$ is the total resistance of the system times G, and we have supposed the mean position of the interface (z = o) to be midway between the ends. $\mathcal{R}$ becomes a function of ρ_α/ρ as well, $\mathcal{R} = \rho h_\alpha(t, t/S, \rho_\alpha/\rho)$. We can state the following about the function h_α

$$h_\alpha(t, \frac{t}{S}, o) = g(t, t/S)$$

$$h_\alpha(t^*, o, \frac{\rho_\alpha}{\rho}) = C \left(\frac{\rho_\alpha}{\rho}\right)^{y_\alpha}$$

$$h_\alpha(t, \frac{t}{S}, \frac{\rho_\alpha}{\rho}) \to (1 - \frac{\rho_\alpha}{\rho})t, \ \frac{t}{L} \to \infty, \ \frac{t}{Z} \to o. \tag{9}$$

Finally, if $\rho_\alpha \neq o$, α = A and B, we must define $\mathcal{R}$ through

$$\mathcal{R} \equiv \mathcal{R}_{\text{Total}} - \frac{1}{2} (\rho_A + \rho_B) Z \tag{10}$$

$\mathcal{R}$ is now given by

$$\mathcal{R} = \rho j(t, \frac{t}{S}, \frac{\rho_A}{\rho}, \xi) \tag{11}$$

where

$$\xi \equiv \rho_B/\rho_A \tag{12}$$

and

$$j(t, \frac{t}{S}, \frac{\rho_A}{\rho}, o) = h_A(t, \frac{t}{S}, \frac{\rho_A}{\rho})$$
$$j(t, \frac{t}{S}, o, \infty) = h_B(t, \frac{t}{S}, \frac{\rho_B}{\rho}) \quad (13)$$

The above statements about R follow straightforwardly from the usual scaling arguments applied in simpler situations.[6,7] Similar results can be set down for the frequency dependence of R and for the interface capacitance and its frequency dependence.[8]

Finally, we note that the exhaustion layer acts as a nonlinear element in regard to both the in-phase and the out-of-phase components of the current relative to the voltage. We therefore restrict consideration to the response of the system to a small ac voltage superposed on an arbitrary dc voltage. The ac resistance and capacitance of the exhaustion layer can therefore be treated as linear elements the values of which depend on the dc bias voltage. For example, our previous experimental results show[9] that the low-frequency capacitance and the current-voltage relation are of Mott-Schottky and diode form respectively, away from the apparent percolation threshold. Therefore, the value of ρ to be inserted into the above formalism is exponentially dependent on bias voltage, and the corresponding value of the local capacitance per unit area obtained from the Mott-Schottky formula. The thickness t is proportional to the square root of the difference between the bias voltage and the flatband potential. Thus, in as much as there is a percolation threshold t^* in the corresponding idealized percolation problem, there is a percolation voltage V^* at which t reaches t^*.

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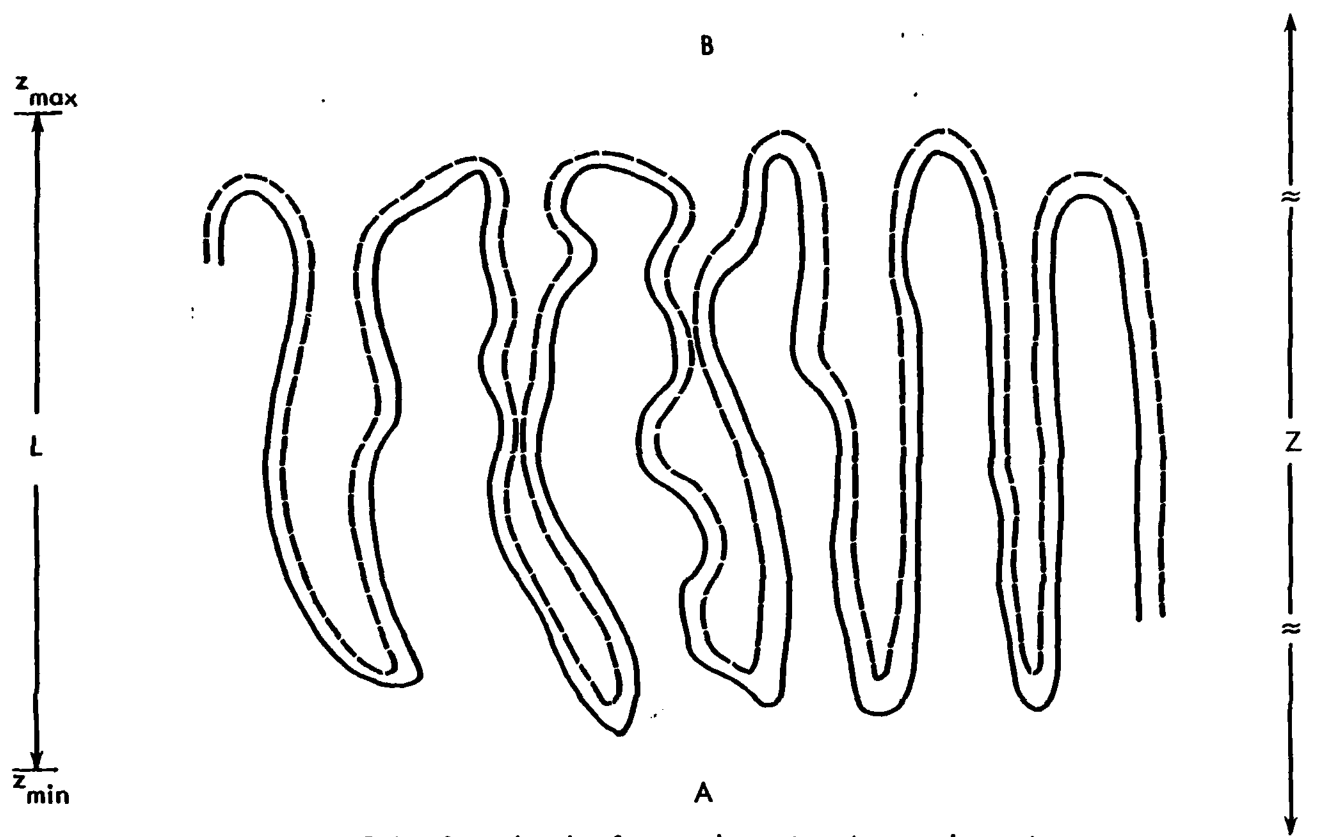

FIGURE 1. Sketch of a rough semiconductor electrode. The symbols are defined in the text.

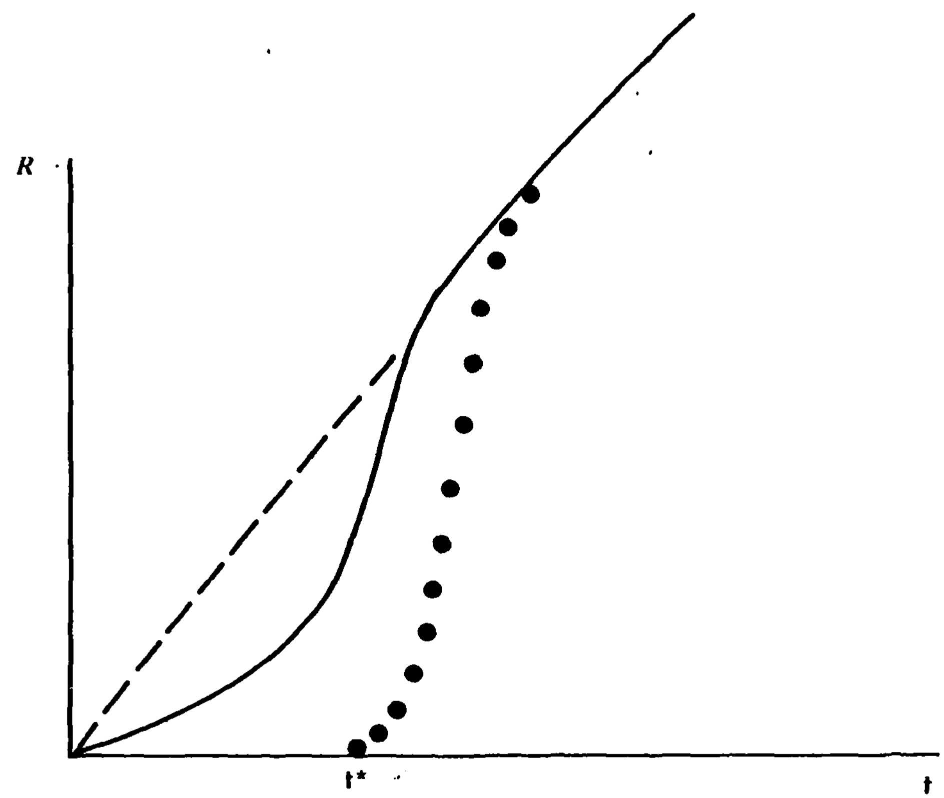

FIGURE 2. Sketch of reduced resistance R versus thickness t of the resistance layer at the interface. The dashed curve corresponds to the smooth interface, the solid curve to a rough interface, and the dotted curve to an infinitely rough interface with a percolation threshold at t^*.

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